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Occupational health risks and fate of chromium in sewage treatment co-treating tannery effluent

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ABSTRACT

This study investigated chromium (Cr) concentrations and fate across processes at a sewage treatment plant (STP) co-treating tannery effluent and assessed health risks for STP workers and farmers using the effluent for irrigation. Two novel secondary treatments, an IPC membrane and a modified constructed wetland, were evaluated as a mitigation method. Methods combined key informant interviews ($n = 4$), observations ($n = 4$), Cr analyses ($n = 132$) and a mass balance approach to track the fate of Cr. Total Cr in the STP influent ranged from 2.49 to 14.95 mg/L, while hexavalent Cr was <100µg/L. Primary sludge contained high total Cr (8,672 to 17,525 mg/kg_{DM}) and hexavalent Cr (0.07 to 0.67 mg/kg_{DM}). Final effluents from the novel and conventional treatments met the Indian discharge standards for total Cr (2 mg/L) and hexavalent Cr (0.1 mg/L). Risk assessment identified three high-risk activities for STP workers, all related to skin contact during primary sludge handling. Farmers irrigated with mixed STP and common effluent treatment plant (CETP) effluent ;the Cr in the CETP effluent was 16 times higher than the STP effluent. The novel technologies did not lower the risks, as they did not address sludge management or CETP effluent quality. Targeted controls for sludge management and coordinated action on CETP discharges are therefore critical.

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1. Introduction

One of India's major industries is leather processing (known as tanning). There are approximately 3,000 tanneries located along the Ganga River (Dwivedi et al. 2018). A cluster of around 400 tanneries can be found in Jajmau, Kanpur, and they discharge around 26 million liters per day (MLD) (26,000 m^3/d) of industrial wastewater (Bassi et al. 2019). The tanning process uses trivalent chromium sulfate to stabilize the leather (Chaudhary et al. 2017; Kokkinos et al. 2019). Trivalent chromium (Cr(III)) can be

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converted to the more toxic hexavalent chromium (Cr(VI)) in the presence of oxidizing agents (Dhal et al. 2013). In occupational settings, chromium (Cr) exposure primarily occurs through ingestion, inhalation, and skin contact (Cheremisinoff and Rosenfeld 2010; Shin et al. 2023). Cr(VI) is both highly toxic and carcinogenic and can lead to allergic dermatitis (ATSDR 2012), skin and nasal irritation, and ulceration upon contact (Shelnutt et al. 2007). While Cr(III) has historically been considered less toxic than Cr(VI), recent studies have linked occupational exposure in tanneries with adverse health effects such as genotoxicity, skin disorders, and immune system alterations (Medeiros et al. 2003; Al Hossain et al. 2019; Islam et al. 2019).

The increase in the number of industries in low- and middle-income countries has resulted in the discharge of untreated industrial effluents into sewers, as in Kanpur (Singh et al. 2009). The fate of Cr in sewage or wastewater is not well understood, largely because municipal wastewater typically contains only low concentrations, ranging from 0.01 to 0.04 mg/L (Henze et al., 2002). This poses several issues, as the level and speciation of Cr are not known and there are no regulations that require the monitoring of Cr in the influent, effluent, or sludges of Indian sewage treatment plants (STPs). This means that occupational health and environmental hazards are unknown, and this is critical when the sludge and effluent are being reused.

In STPs, the fate of Cr depends on the treatment processes and the chemical form of the Cr. Cr(III) tends to associate with sludge, while Cr(VI) is more soluble and may remain in the water phase if not reduced to Cr(III) (Gorny et al. 2016). In STPs Cr(III) primarily causes surface-level inhibition by binding to the active sites of microbial cellular membranes, which reduces the capacity for organic matter uptake (Vaiopoulou and Gikas 2012). In contrast, Cr(VI) can enter microbial cells and disrupt microbial respiration (Vaiopoulou and Gikas 2012; Viti et al. 2014). This dual effect reduces the organic matter adsorption capacity and impacts the COD removal efficiency in activated sludge systems (Vanková et al. 1999). Studies show that lethal concentrations of Cr(III) for activated sludge are around 160 mg/L, while levels between 40 and 49 mg/L inhibit aerobic microbial respiration by 50% (Wong et al. 1997; Vanková et al. 1999; Gikas and Romanos 2006). Additionally, the presence of Cr in sewage sludge can result in environmental pollution through disposal (Wang et al. 2005; Fuentes et al. 2006). Even if the main form of Cr is Cr(III) , it can be converted in the soil to Cr(VI) *via* oxidizing agents such as manganese, lead oxides and high pH (Dhal et al. 2013), this form is more toxic and environmentally mobile.

In India, 32 of 54 cities with a population of more than 1 million have adopted wastewater reuse projects, and 17% of the wastewater generated from these cities is being recycled and reused (CPHEEO 2021). The use of treated wastewater for irrigation poses challenges regarding water quality, risk mitigation and monitoring of treatment performance (Drechsel et al. 2022). Occupational health risks for wastewater or sludge re-users typically focus on potential infections due to pathogens in the effluent or sludge, which pose short-term biological risks. The presence of chemical compounds through informal industrial discharges in low- and middle-income countries makes it necessary to evaluate the long-term chemical risks (Lazarova et al. 2004). Wastewater reuse guidelines focus primarily on biological risks, while chemical risks, such as heavy metals from informal discharges, are often overlooked (Shoushtarian and Negahban-

Azar 2020). Due to the increased use of wastewater in India, the 'National Framework on Safe Reuse of Treated Water' and the 'Guidelines for Reuse of Treated Sewage in Reference to the Item of Circular Economy' have proposed standards for the reuse of municipal wastewater, including organic and microbial parameters (MJS 2022; CPCB 2024), but inorganic chemicals such as Cr are not included due to their low concentrations in municipal wastewater (Henze et al. 2002). There are standards for wastewater treatment plants treating industrial effluent known as common effluent treatment plants (CETPs) and the standard for total Cr is 2.0 mg/L for use in irrigation or discharge to land; and 0.1 mg/L for Cr(VI) (MoEFCC, 2016). Currently, there are no regulatory standards for Cr in municipal sludge in India, but the maximum concentration for organic compost or phosphorous-rich organic manure is 50 mg $Cr/kg_{dry\ matter(DM)}$ (M/o Agriculture 2013).

As the informal discharge of industrial effluents into sewers is largely ignored, past occupational health risk assessment studies on wastewater have concentrated on microbiological risk (Ensink et al. 2008; Jackson and Vuong 2014; Babalola et al. 2023). In India, they have explored nematode infection risk (Ensink et al. 2008), microbiological and physical risks (Babalola et al. 2023) for farmers using treated effluents. No studies were found that have explored chemical occupational health risks related to the co-treatment of municipal and industrial wastewater. Therefore, this study aimed to explore the level and fate of Cr and its speciation across the different processes at a STP co-treating municipal and tannery effluent in Kanpur, India. The study also assessed the occupational health risks of Cr for STP workers and farmers using effluent to irrigate crops. Additionally, the study explored the use of novel secondary treatment technologies (membrane filtration and modified constructed wetlands) as a mitigation measure for occupational health risks.

2. Methodology

2.1. Study background

This study was part of the Pavitra Ganga Project, a research and innovation initiative from the European Union/India Cooperation that aimed to tackle the pollution of the Ganga River through the implementation of innovative wastewater treatment technologies and resource recovery opportunities in Kanpur and Delhi. The Pavitra Ganga project tested innovative technologies as alternative secondary treatments at the demonstration site in Kanpur (Pavitra Ganga 2020). These technologies included a combination of an integrated permeate channel (IPC) membrane and a modified constructed wetland (CW+). The aim of the IPC membrane was to produce permeate with no suspended solids or coliforms, while the CW+ aimed to enhance heavy metal removal from wastewater by integrating vertical flow constructed wetlands with sorbents like granular activated carbon (UN-HABITAT 2008; Pavitra Ganga Project 2023). The purpose of these technologies was to produce higher-quality effluent for reuse. The present study focuses on semiquantitative risk assessment for STP workers and farmers, while environmental risks are investigated in a related project in the same case study area. The present study was undertaken between July and August 2023, during the monsoon season.

2.2. Case study area

Kanpur City, located on the southern bank of the Ganga River and is the industrial and commercial capital of Uttar Pradesh due to its leather industry, which contributes to around 13.5% of the country's leather exports (JPS Associates LTD 2006). The city's population is 2.9 million and has tripled in the last three decades (Bassi et al. 2019). The effluent produced at the tanneries located in the Jajmau (a suburb of Kanpur) has increased from 9 MLD (9,000 m^3/d) in 1994 to 26 MLD (26,000 m^3/d) in 2015 (Bassi et al. 2019). There are three operational wastewater treatment plants in Jajmau: two STPs designed to treat municipal wastewater, one designed to treat 130 MLD (130,000 m^3/d) and one designed to treat 5 MLD (5,000 m^3/d); and one CETP designed to treat 36 MLD (36,000 m^3/d) of combined tannery and municipal wastewater (Babalola et al. 2023).

This study focused on the 130 MLD STP at Jajmau, which uses an activated sludge process (ASP). This STP was commissioned in 1999 and currently treats 100 MLD (100,000 m^3/d) (CPCB 2021). Informal effluent discharges from tanneries enter the Jajmau STP *via* discharge into the sewers. This has negatively affected the STP performance, especially as the number of tanneries in Jajmau has increased (JNNURM 2006). The effluent from Jajmau STP is combined with the effluent of the CETP and discharged into a concrete irrigation channel that supplies irrigation water to 2,500 ha of farmland in nearby villages (JNNURM 2006). Around 40 villages receive the treated wastewater through these irrigation channels. The wastewater reuse scheme has been implemented for 37 years and the villages have received mixed effluent for 24 years (JPS Associates LTD 2006). They use flood irrigation to farm rice, wheat and millet (Babalola et al. 2023).

2.3. Sampling and Cr determination

Samples were taken at 11 sampling points (S1-S11) along the treatment train and reuse scheme (Figure 1) during three campaigns. For all wastewaters, composite samples were taken at intervals of two hours over an 8-h period (10 am, 12 pm, 2 pm, 4 pm). No precipitation was recorded during the sampling campaigns. Each composite sample was collected in duplicate, resulting in two samples per sample point. Separate samples were taken for total Cr and Cr(VI) determinations. Per campaign, this generated 22 samples per analyte and 44 samples in total. A total of 132 samples were collected and analyzed over the three sampling campaigns. All composite samples were collected in 250 mL high-density polyethylene (HDPE) containers and transported to the laboratory in a cool box to maintain a temperature of $\leq 6\,^{\circ}C$ until analysis (Baird and Bridgewater 2017). As the most common Cr oxidation states are Cr(III) and Cr(VI), total Cr represents the sum of these two oxidation states. Unfiltered samples were analyzed for total Cr and Cr(VI). Cr(III) was calculated from the total Cr minus Cr(VI). This was done to explore the speciation of Cr in wastewaters and sludges and to detect any chemical transformation in the STP and due to the different health risks related to the different forms of Cr.

The sample preparation followed the EPA methods SW-846 Method 3060 A and SW-846 Test Method 6020B (Baird and Bridgewater 2017). The Cr(VI) analysis was undertaken using the colorimetric method with diphenylcarbazide (3500-Cr B) (Baird and

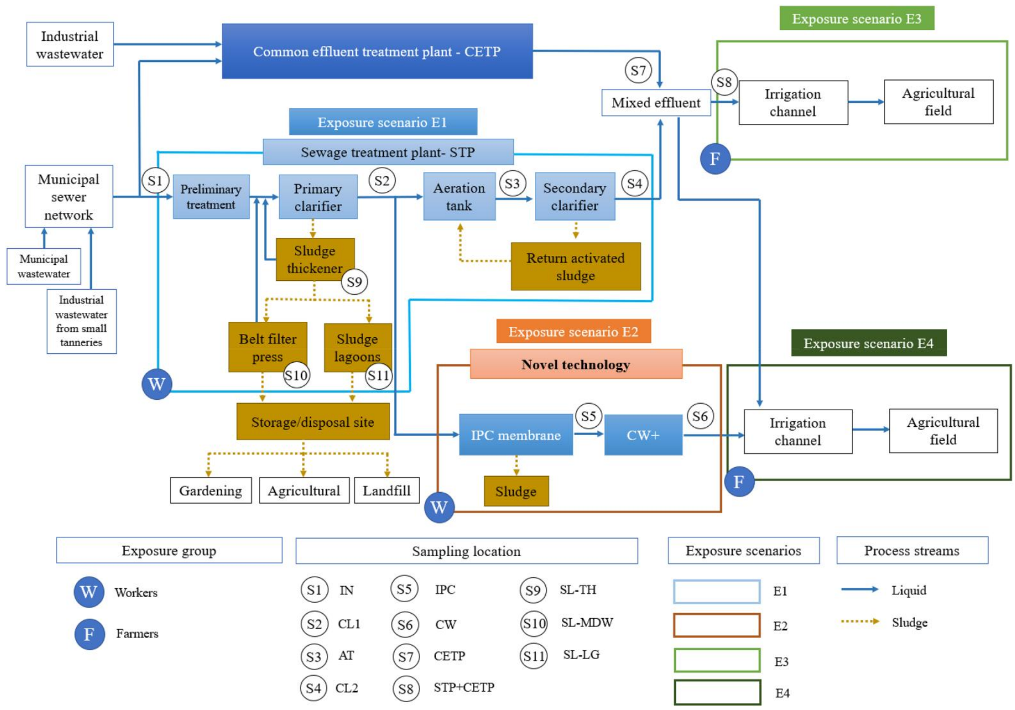

Figure 1. Process flow for the semi-quantitative health risk assessment.
Note: IN: plant influent, CL1: primary clarifier, AT: aeration tank, CL2: secondary clarifier, IPC: integrated permeated channel, CW: constructed wetland, CETP: common effluent treatment plant effluent, SPT+CETP: mixed effluent from STP and CETP, SL-TH: sludge thickener, SL-MDW: mechanical dewatered sludge, SL-LG: sludge from sludge lagoons.

Bridgewater 2017). APHA method 3030E (acid digestion with HNO_3 conc.) was used for total Cr determination, which was done using an ICP-MS Agilent 7800 (Baird and Bridgewater 2017). As there are Indian Cr standard for reuse, the concentrations were assessed in accordance with standards for discharge of treated effluent from CETPs in India which is 2.0 mg/L for total Cr and 0.1 mg/L Cr(VI) (MoEFCC, 2016), following the wastewater parameters by the Central Pollution Control Board (CPCB 2011).

Duplicate grab samples of thickened sludge (SL-TH; S9), mechanically dewatered sludge (SL-MDW; S10), and sludge from lagoons (SL-LG; S11) were collected at 10 am and 4 pm. The two samples were homogenized to form a composite sample. Transport conditions were identical to those used for wastewater samples. The samples were dried in an oven at 105 °C for 48 h before digestion andanalyzed according to methods EPA 3050B and APHA 3500CrB (U.S. EPA 1996; Baird and Bridgewater 2017) in Vimta Labs, located in Hyderabad, India (CPCB-recognized environmental lab). The Cr concentrations were assessed and compared with the maximum concentration limit for organic compost and phosphorous-rich organic manure (M/o Agriculture 2013).

2.4. Cr mass balance

For the process mapping (Figure 1) and the Cr mass balances, data were obtained from key informant interviews (KIIs). The KIIs ($n = 4$) were conducted to collect data on the flow rates, sludge volume or mass, and sludge fate. The interviews were face-to-face, notes were taken, and the data was summarized and analyzed. Data from KIIs was triangulated with secondary data from the Pavitra Ganga project as well as journal articles, manuals, and reports related to the study. The measured concentrations of total Cr and Cr(VI) were multiplied by the process flow rates and sludge quantities in each treatment step to obtain the Cr mass balance (Table S.1 Supplementary Materials).

2.5. Occupational health risk assessment

A semi-quantitative risk assessment framework (SQRA) was adopted in the study for two main reasons: it allowed for a structured estimation of Cr-related risks using measured concentrations and exposure scenario assumptions, and it was better suited to the urban, resource-limited setting of the study, where detailed exposure data were lacking. The WHO Sanitation Safety Planning (SSP) framework was used for its practical applicability in such contexts, following the manual for guidance (WHO 2016; Domini et al. 2017; Frattarola et al. 2019). The system boundary for this study comprises Jajmau STP, the novel technologies IPC membrane and CW+ (Figure 1) and the reuse of treated wastewater for irrigation in the villages.

The hazardous events considered for the occupational risk assessment were those related to Cr exposure pathways: skin contact, inhalation, and accidental ingestion. The two exposure groups were the STP workers and farmers. The preliminary system boundary of Babalola et al. (2023) was expanded to include sludge processes, which were used to identify the hazardous events for the exposure groups (Figure 1). KIIs with the workers ($n = 2$) in the STP and observation during field visits were used to identify hazards and hazardous events. The soil exposure pathway, including potential Cr accumulation and crop uptake, was not assessed in this study.

Table 1. Cr effluent quality standards and minimal risk levels (MRL) for skin contact, inhalation and ingestion exposure.

	MOEFCC[1] effluent discharge standards for tanneries	ATSDR[2] MRL inhalation	ATSDR[2] MRL ingestion	ATSDR[2] MRL skin contact
Cr(III)	None	1×10^{-4} mg/m^3 (lung inflammations, nasal and larynx lesions)	*33 – 74 mg/kg/d (reproductive effects)	*1 mg /L (Cr induced allergic dermatitis)
Cr(VI)	0.1 mg/L	5×10^{-6} mg/m^3 (nasal irritation, mucosal atrophy, impaired lung function)	9×10^{-4} mg/kg/d (hyperplasia of duodenum)	*4–25 mg/L (Cr induced allergic dermatitis)
Total Cr	2 mg/L	None	None	None

[1]MoEFCC (2016); [2]ATSDR (2012). The lower MRLs are displayed for intermediate and chronic effects. *Insufficient data to derive MRL, concentrations showing effects indicated from literature (ATSDR 2012).

Table 2.Severity scale developed in this study for Cr exposure and potential health effects (aligned to severity categories in (WHO 2016).

Severity (S)		Inhalation	Skin contact	Ingestion
1	Insignificant Cr(III)/Cr(VI) concentrations far below MRLs for skin contact, ingestion or inhalation exposure resulting in no or negligible health effects.	Cr(III) $< 4 \times 10^{-6}$ mg/m^3 Cr(VI) $< 5 \times 10^{-8}$ mg/m^3	Cr(III) < 0.01 mg/L Cr(VI) < 0.04 mg/L	Cr(III) < 0.023 mg/d for a 70 kg person ingesting a drop (0.002 L)
2	Minor Cr(III)/Cr(VI) concentrations below MRLs for skin contact, ingestion or inhalation potentially exposure resulting in minor health effects.	4×10^{-6} mg/m^3 < Cr(III) $< 4 \times 10^{-4}$ mg/m^3 5×10^{-8} mg/m^3 < Cr(VI) $< 5 \times 10^{-6}$ mg/m^3	0.01 mg/L < Cr(III) < 1 mg/L 0.04 mg/L < Cr(VI) < 4 mg/L	0.023 mg/d < Cr(III) < 2.310 mg/d for a 70 kg person ingesting a drop (0.002 L)
4	Moderate Cr(III)/Cr(VI) concentrations within the range of the MRLs skin contact, ingestion or inhalation exposure potentially resulting in self-limiting health effects or minor illness.	4×10^{-4} mg/m^3 < Cr(III) < 0.04 mg/m^3 5×10^{-6} mg/m^3 < Cr(VI) $< 5 \times 10^{-4}$ mg/m^3	1 mg/L < Cr(III) < 1 g/L 4 mg/L < Cr(VI)< 25 mg/L	Cr(III) > 2.310 mg/d for a 70 kg person ingesting a drop (0.002 L)
8	Major Cr(III)/Cr(VI) concentrations above the range of the MRLs for skin contact, ingestion or inhalation exposure potentially resulting in illness.	Cr(III) > 0.04 mg/m^3; Cr(VI) $> 5 \times 10^{-4}$ mg/m^3	Cr(III) > 1 g/L; Cr(VI) > 25 mg/L	**Absence of data showing adverse effects of chronic oral exposure*
16	Catastrophic Cr(III)/Cr(VI) concentrations far above the range of the MRLs for skin contact, ingestion or inhalation exposure, potentially resulting in serious illness	**Insufficient data to derive acute MRL*	**Insufficient data to derive acute MRL*	**Insufficient data to derive acute MRL*

*Insufficient data to derive acute toxicity MRL (ATSDR 2012).

The likelihood was assessed according to the likelihood scale (WHO 2016) (Table S.2 Supplementary Materials). The severity of Cr exposure was assessed with an adapted severity scale related to the Cr level at the different sampling points along the treatment train and the minimal risk levels (MRL) for inhalation, ingestion, and dermal efflect levels for skin contact (Table 1). The risk score was determined by multiplying the likelihood score by the severity score and it was used to categorize the hazards into four risk levels: low, medium, high and very high (Table S.3 Supplementary Materials).

Table 2 (ATSDR 2012; WHO 2016).The severity scale used in this study was derived from the minimum risk levels (MRLs) (Table 1)and the severity categories in WHO (2016) and is presented in Table 2 (ATSDR 2012; WHO 2016). Severity categories were defined relative to published MRLs for Cr(III) and Cr(VI), following conventions used in established semi-quantitative risk assessment frameworks, such as the WHO IPCS chemical risk assessment guidance (IPCS 2004). Specifically, "below MRL" refers to concentrations lower than health-based benchmarks and is considered unlikely to result in adverse health effects. "Within the range of the MRL" includes concentrations near or up to the benchmark value, representing potential for minor or self-limiting effects. "Above MRL" denotes concentrations exceeding the MRL by up to approximately one order of magnitude ($\leq 10\times$), where health effects may become more likely. "Far above MRL" refers to concentrations greater than one order of magnitude above the MRL ($>10\times$), indicating a substantially elevated risk of adverse health outcomes. The use of MRLs and related health-based thresholds to characterize risk is consistent with IPCS chemical risk assessment guidance (IPCS, 2004), where such benchmarks help determine the likelihood and magnitude of adverse health effects based on exposure. While frameworks like WHO's Sanitation Safety Planning (WHO 2016) define severity qualitatively based on potential health outcomes, the IPCS approach incorporates quantitative comparisons of exposure to toxicological reference values. This is particularly suitable for chemical hazards such as Cr(III) and Cr(VI), where benchmark-based methods can inform both risk estimation and prioritization.

3. Results

The system boundaries are shown in Figure 1, along with the exposure groups, the sampling points, and the exposure scenarios. The exposure scenario for STP workers included conventional treatment (E1) and novel technologies (E2). The exposure scenario for farmers included the irrigation using treated wastewater from conventional treatment + CETP (E3) and novel technologies + CETP (E4).

The Jajmau STP treatment train starts with a screen chamber and grit removal as a preliminary treatment to remove the coarse debris and floating materials (Figure 1) (Babalola et al. 2023). The influent is then divided over three parallel primary clarifiers, each with a capacity of 43 MLD (43,000 m^3/d) (KII-01). In this step, the particulate matter settles and is removed from the liquid stream. The secondary treatment used is an ASP system, and finally, there are three secondary clarifiers for the liquid stream (Babalola et al. 2023). The pilot IPC membrane receives 7 m^3/d of effluent from the primary clarifier (KII-03). The permeate flows into the CW+ that treats around 0.75 m^3/d (KII-04).

Several processes are used for sludge management at Jajmau STP: gravity thickeners, mechanical dewatering, drying beds and sludge lagoons (KII-01). Under normal conditions, primary sludge is thickened and mixed with secondary sludge. According to KII-01, 75-80% of the activated sludge is returned to the aeration tank, and 20-25% is discarded. However, all activated sludge was returned to the aeration tank during the study, so only primary sludge was produced. Two belt filter presses operate continuously. There are 38 drying beds in the STP with an approximate area of 400 m^2 each, which were not in operation during the study period, as it was in the monsoon season.

During this study, only the sludge lagoons and the belt filter presses were operational and assessed for occupational health risks.

The sludge was transported from Jajmau STP to a storage/disposal site managed by the Kanpur Nagar Nigam (Kanpur Municipal Corporation), where it is reused as fertilizer for gardening activities and by farmers who do not use the mixed STP-CETP effluent for irrigation (KII-01). Hence, sludge reuse among farmers in the study area was not considered in the SQRA.

3.1. Concentrations of total Cr, Cr (VI) and Cr (III)

Total Cr and Cr(VI) concentrations are shown in Table 3. Total Cr concentrations in the wastewater samples ranged from 0.03 to 57.52 mg/L with the lowest and highest levels being measured at sampling points S5/S6 and S3, respectively. All wastewater samples (S1-S8, Table 3) were below the limit of detection for Cr(VI), which is 100 µg/L (Baird and Bridgewater 2017). Thus, the wastewater quality complies with the Indian effluent treatment standard for CETPs and tanneries 0.1 mg/L of Cr(VI) (MoEFCC, 2016). Since Cr(VI) concentration in wastewater was below detection limit, all Cr occurs as Cr(III). Total Cr concentrations in the sludge after the belt filter press were extremely high and ranged between 8,672 to 17,525 mg of total Cr/kg_{DM} (S10, Table 3). Cr(VI) concentrations in sludge ranged from 0.07 to 0.67 mg/kg_{DM}.

3.2. Cr mass balance

Daily mass flow rates of Cr in the STP process streams are shown in Figure 2. Cr variation in the process stream of novel technologies is shown in Figure 3. The mean daily mass of total Cr entering the plant in the influent stream was 764 kg/d. The primary clarifier removed 617 kg/d (Figure 2), which represents an 80.8% reduction of the total Cr mass. As shown by Table 3 (S9-S11) total Cr accumulated in the primary sludge. The daily mass of total Cr leaving the system in the effluent was 20 kg/d (Figure 2), so the overall removal of total Cr by the STP was 97.4%. The complete recirculation of the activated sludge in the ASP resulted in an accumulation of 127 kg/d (Figure 2).

Regarding the novel technologies, due to the pilot volume, the influent of the IPC membrane contained 10 g total Cr/d (Figure 3). In comparison, the effluent only contained 0.25 g total Cr/d; a 97% total Cr reduction was achieved. As this is a secondary process, it is important to note that most of the removal takes place during the primary treatment. The Cr reduction in the CW+ was 90%. As with the standard system, most of the total Cr ends up in the sludge of the system, and less than 1% of total Cr leaves the system in the effluent.

3.3. Occupational health risk assessment

3.3.1. Occupational health risk assessment for STP workers related to the conventional treatment

The health risk assessment was conducted for the STP workers of the conventional treatment (E1, Figure 1). Hazardous events are mainly related to exposure to untreated

Table 3. Total Cr concentrations at different sampling points in STP Jajmau.

		First campaign		Second campaign		Third campaign	
Sampling point	Sample	Total Cr (mg/L) ($n = 2$)	Cr(VI) ($n = 2$)	Total Cr (mg/L) ($n = 2$)	Cr(VI) ($n = 2$)	Total Cr (mg/L) ($n = 2$)	Cr(VI) ($n = 2$)
S1	IN	13.52−14.95	below 100 µg/L	2.49−2.70	below 100 µg/L	4.78−5.21	below 100 µg/L
S2	CL1	1.73−2.13		0.68−0.86		1.49−1.52	
S3	AT	43.51−45.68		30.77−57.52		15.66−22.17	
S4	CL2	0.20−0.22		0.13		0.23−0.24	
S5	IPC	0.04−0.05		0.03−0.04		0.03	
S6	CW+	0.04		0.03		0.03	
S7	CETP	2.98		2.97−3.56		3.06−3.49	
S8	STP+CETP	1.34−1.58		0.78−0.95		0.28−2.09	
S9	SL-TH (mg/kg)	14,913*	0.24*	19,136*	0.07*	15,604*	0.39*
S10	SL-MDW (mg/kg)	8,672*	0.67*	17,525*	0.49*	14,088*	0.4*
S11	SL-LG (mg/kg)	15,045*	0.28*	16,296*	0.52*	16,532*	0.63*

Note: *units mg/kg_{DM}. IN: plant influent; CL1: primary clarifier; AT: aeration tank; CL2: secondary clarifier; IPC: integrated permeated channel; CW: constructed wetland; CETP: common effluent treatment plant effluent; SPT+CETP: mixed effluent from STP and CETP; SL-TH: sludge thickener; SL-MDW: mechanical dewatered sludge; SL-LG: sludge from sludge lagoons.

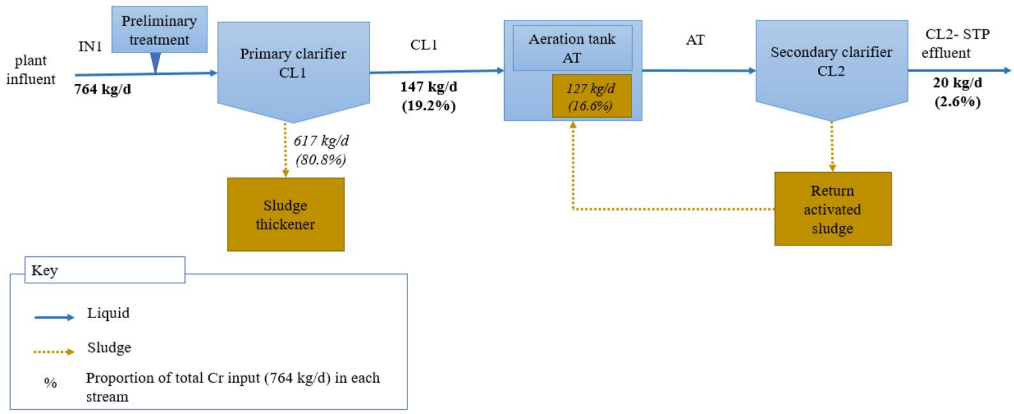

Figure 2. Mass balance diagram showing total Cr flows at different stages of the process streams at STP Jajmau. Note: Measured values are in bold, and calculated values are in italics.

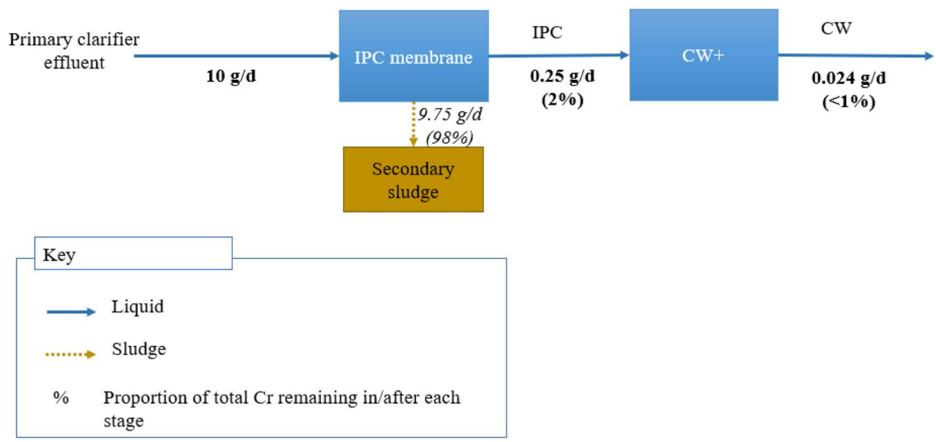

Figure 3. Massbalance diagram showing Cr flows in the process streams in the novel technologies. Note: Measured values are in bold, and calculated values are in italics.

and partially treated sewage during operation and maintenance, which is present throughout the whole treatment train. However, due to the low concentrations of Cr(VI) in the wastewater (Table 3), the risk assessment considered only Cr(III) concentrations, except in the sludge management processes (S9-S11, Table 3). Ten hazards were identified for the STP workers based on the observations, interviews with the workers and the activities identified in a previous study (Babalola et al. 2023).

All likelihood, severity and risk scores are given in the Supplementary Materials Table S. 4–S.6. A summary of the risk scores across the treatment trains can be seen in Figure 4.

The preliminary treatment included operational activities such as collecting the debris from the screen chamber and cleaning the machines involving the exposure to Cr-containing materials coming from the tanneries, such as flesh and hair (Puhazhselvan et al. 2022). During primary treatment, workers are exposed to untreated sewage during manual removal of scum from the primary clarifiers. For secondary treatment, aerosols containing Cr(III) are generated. The mentioned activities entail medium occupational risks (Figure 4, Table S.4 Supplementary Materials).

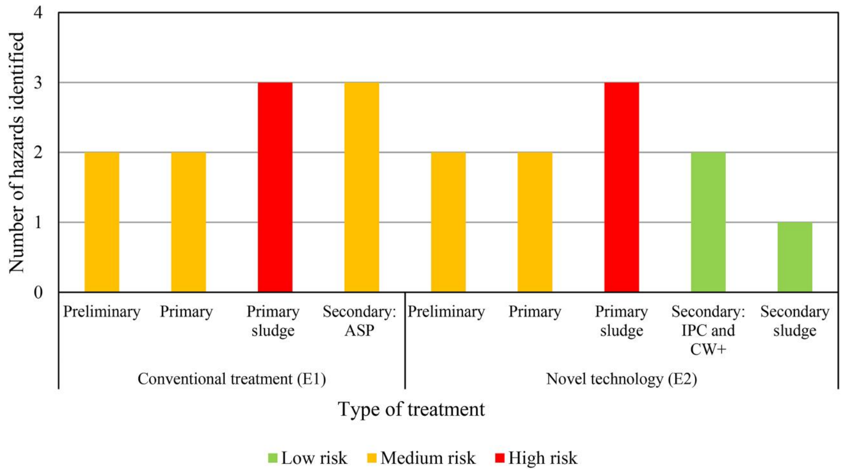

Figure 4. Number of hazards and risk levels for STP workers using conventional treatment and novel technologies.

Inhalation exposure of 4×10^{-4} mg Cr(III)/m^3 and 5×10^{-6} mg Cr(VI)/m^3 are classified as minimum risk levels (Table 1). Annual average concentrations of total Cr of 1.08×10^{-5} mg/m^3 has been found in previous studies around an aeration tank, which is 40 times less than the MRL for Cr(III) (Yang et al. 2020). Thus, inhalation of aerosols is related to minor health outcomes in this study (severity level = 2, in Table 2). As skin contact exposure to Cr(III) in wastewater or sludge at concentrations of > 1 mg/L has been associated with allergic dermatitis in sensitized individuals (Table 1) any handling of wastewater and sludge throughout the STP that exceeds this level was considered as moderate severity(level 4; Table 2).

The MRL of Cr(III) for ingestion is 33–74 mg/kg/d for gastrointestinal effects (Table 1). If 0.002 L of effluent from the aeration tank (mean concentration S3, Table 3) was accidentally ingested by a 70 kg person, ca. 0.07 mg Cr(III) is consumed, which is 32 times less than the daily MRL (i.e., 2.310 mg, Table 2), if it happens only once per day. Therefore, accidental ingestion of wastewater and sludge containing Cr(III) has been classified as minor health effects (severity level = 2, Table 2).

In managing sludge, various treatment steps involved direct worker contact. Typically, one worker oversees operations during thickening and dewatering without direct sludge contact. However, “sweepers" were directly involved in removing debris or materials from pipes, exposing them to sludge for at least 30 min daily (KII-02). Observations indicated potential hazards during sludge thickening, such as debris removal and direct sludge contact during belt filter press cleaning in mechanical dewatering. While there were no reported long-term skin issues, workers handling sludge experienced a burning sensation on their skin that persisted for some time (KII-02). Sludge management processes had a risk score of 24 related to Cr(III) and Cr(VI) exposure, all indicating a high-risk (Figure 4, Table S.4 and S.5 Supplementary Materials), considering genotoxic effects, dermatological conditions and immune system disturbances associated to workers exposed to Cr(III) (Medeiros et al. 2003; Al Hossain et al. 2019; Islam et al. 2019).

The existing control measures within the STP included using personal protective equipment (PPE): gloves, safety boots, helmets, and safety belts. Although workers were instructed to use PPE, they were not always provided (Babalola et al. 2023). During the observations, workers were observed without any PPE, leaving their hands and legs exposed to untreated sewage, effluents and sludges.

3.3.2. Occupational health risk assessment for STP workers related to the novel technologies

The boundaries of E2 can be found in Figure 1. If the novel technologies were to be implemented, the risk related to the preliminary and primary treatment, as well as primary sludge management, would remain the same as the conventional treatment, so the hazardous events, hazards, and risk levels would remain the same (E2, Figure 4).

Inhalation and accidental ingestion were not considered for the novel secondary technology as no aerosols are formed, and the enclosed IPC system minimizes worker contact with the influent and effluent in activities like routine maintenance (Babalola et al. 2023). As the Cr(III) concentration was, on average, 0.04 mg/L (S5, Table 1), the severity of skin contact exposure was minor (Table 2), and the risk score was low (Table S.6 Supplementary Materials). The IPC membrane technology would produce less sludge than conventional treatment, reducing the workers' sludge handling frequency.

The CW+ requires no contact of the workers, and the operation and maintenance mostly involved adjusting the influent level and visual inspection of walls, inlets, outlets and vegetation (UN-HABITAT 2008). Eventually, the substrate will need to be changed when it becomes saturated. Case studies on vertical flow wetlands suggest that this frequency varies from 5 to 10 years (UN-HABITAT 2008). Nevertheless, the mean total Cr concentration in the inlet was around 0.04 mg/L (Table S.1 Supplementary Materials), which was considered as minor severity for skin contact (Table 2).

3.3.3. Occupational health risk assessment for farmers

Due to the use of flood irrigation, the formation of aerosols and accidental ingestion were not considered hazards. However, exposure to Cr-containing treated wastewater through skin contact was considered during flood irrigation and farming activities. A previous study reported no use of facemasks, gloves or boots as protective equipment during farming activities, so the farmers were continuously in direct contact with the irrigation water (Babalola et al. 2023).

The concentration of total Cr and, therefore, Cr(III) in the mixed effluent (S8, Table 3) was above the MRL for skin contact exposure to Cr(III) (Table 1), so the risk score for this activity was 20 (high-risk) (Table S.7 Supplementary Materials). Cr(VI) was not considered for the exposure scenario since it was below 100 μg/L, hence below the MRL (Table 1). A majority of the total Cr in the irrigation channels originates from the CETP (S7 and S8, Table 3). Hence, most of the risk related to the exposure of farmers to Cr(III) came from CETP effluent. The novel technologies did not mitigate the high risks to STP workers and farmers because they did not address sludge handling or CETP effluent quality.

4. Discussion

As the predominant oxidation state in the wastewater was Cr(III), this indicates Cr(III) is not being converted to Cr(VI). These are positive results as Cr(VI) is more toxic and carcinogenic compared to Cr(III) (ATSDR 2012; Sun and Costa 2022). The concentrations of total Cr (mainly Cr(III)) were highly variable in the influent, ranging from 2.49 mg/L up to 14.95 mg/L (S1, Table 3). These levels are between 64 and 373 times higher than typical levels for municipal wastewater, where Cr concentrations are between 0.01 mg/L and 0.04 mg/L globally (Henze et al. 2002). These finding prove that the tanneries were informally discharging into the municipal sewer network and supports research that found that approximately 30-40% of Cr salts remain in the wastewater in tannery processing (Cassano et al. 1997; Wang et al. 2007).

The variability in the total Cr concentrations in the influent may be due to the study being undertaken during the monsoon season, heavy rains may have diluted the influent in the combined sewers, or there may have been different activities and working patterns in the tanneries during sampling. It should be noted that higher Cr concentrations would be expected during the dry season, as there would be no dilution of the influent. This means that during the dry season the STP workers and farmers may be exposed to higher concentrations of Cr than in the monsoon season, which may increase their risk.

There was a significant reduction (approximately 81%) of total Cr after the primary clarifier (Figure 2), which demonstrating that total Cr and Cr(III) were accumulating in the primary sludge. This was expected as the predominant oxidation state was Cr(III) which associates with particulate matter (Gómez & Callao 2006). The total Cr concentration in the aeration tank (S3, Table 3) was extremely high compared to the concentrations in the secondary clarifier (S4, Table 3). This was due to the recirculation of sludge without any wastage, which resulted in the progressive accumulation of Cr in the biomass. Activated sludge is known to adsorb Cr(III) (Ramteke et al. 2010; Vaiopoulou and Gikas 2012). The maximum total Cr concentration measured in the ASP was 57.52 mg/L; this concentration is known to affect the COD removal efficiency due to the competition between organic matter and heavy metals for binding to the surface of the biomass (Vanková et al. 1999).

There was a 97.4% reduction of total Cr in the effluent from the STP. The concentrations in the effluent (S4, Table 3) are below the CETP effluent discharge standards of 2 mg/L for total Cr (MoEFCC, 2016). The total Cr concentrations in CETP effluent ranged from 2.97 to 3.56 mg/L (S7, Table 3), exceeding the discharge standards for CETPs (MoEFCC, 2016). After mixing of the STP and CETP effluents in a ratio of 65-70% STP to 30-35% CETP effluent (KII-02), the final concentrations of total Cr in the irrigation channel were between 0.28 and 2.09 mg/L (S8, Table 3), this is around the maximum permissible limit for total Cr in CETP effluents (2 mg/L) (MoEFCC, 2016).

The total Cr in sludge were up to 300 times higher than the maximum concentration limit for organic compost and phosphorous-rich organic manure (50 mg Cr/kg_{DM})(M/o Agriculture 2013). Cr(III) levels in all sludges were in the same range (S9 to S11, Table 3). The Cr(VI) was either concentrated in the sludge or the Cr(III) present in the sludge was oxidized to Cr(VI) due to environmental conditions in the lagoons or mechanical dewatering process. This transformation is possible as studies on tannery waste in Kanpur found Cr(VI) in aged sludge, even though the sludge initially contained Cr(III), indicating that Cr(III) was being oxidized to Cr(VI) over time (Apte et al. 2005; Kumar et al. 2023).

The total Cr concentrations in the effluents of the IPC membrane and CW+ were between 0.03 and 0.05 mg/L (S5 and S6, Table 3), complying with the permissible standards for CETPs (2 mg/L) (MoEFCC, 2016). Considering that the novel technology received effluent from CL1 (Figure 1), the total Cr reduction in the IPC was 98%, higher than the reduction achieved by the ASP of 86%. Comparing the total Cr concentrations in the influent of the IPC with the effluent (S2 and S5, Table 3), most of the Cr remained in the residual sludge of the system.

Although there was no wastage of activated sludge during the study period (Figure 1), 25 to 30% of the sludge is wasted under normal operating conditions (KII-01). This means that between 32 to 38 kg/day of Cr would end up in the sludge stream. This increase in Cr concentration by 5 to 6% could raise health risks related to sludge handling. It should also be noted that significant Cr accumulation occurs in the secondary clarifier under normal operating conditions, estimated to be between 95 to 102 kg/day. This accumulation could substantially impact the system's functioning, as previously discussed. This demonstrates the importance of using a mass balance approach, as the points above cannot be fully explored when only concentrations are considered.

It is important to note that even though the CW+ reduced Cr by 90% of the daily mass entering the novel technology (Table 3), most of the removal takes place in the previous step (IPC membrane). The mean concentration of total Cr is already below the maximum permissible limit for CETPs (2 mg/L) before it enters the secondary treatment processes (MoEFCC, 2016). These results have significant implications for a full-scale implementation of the novel technologies and the health risk assessment of the reuse scheme. While the novel technologies are innovative and more efficient for fecal indicator organisms (Babalola et al. 2023), they do not offer any added benefit for Cr removal, as the primary clarifiers in the conventional treatment are already effective.

When the risk scores for both technologies are compared (Figure 4) ten hazards were identified along the treatment train for the conventional treatment (seven medium and three high). For the treatment train containing the novel technologies ten hazards were identified (three low, four medium and three high). The only change was the risks associated with skin contact in the secondary treatment, which changed from medium- to low-risk (Figure 4) when the novel technologies were implemented, and the risks associated with inhalation and ingestion disappeared due to the enclosed system.

Although the risk assessment covered activities conducted throughout the year, the Cr levels used to determine the severity of the risk were based on samples taken during the monsoon season. As previously explained, Cr concentrations in the wastewater may be higher during the dry season, which means the overall exposure risk could be greater than currently estimated and should also be considered when reviewing the data on risks to farmers.

As most of the risk related to the exposure of farmers to Cr(III) came from CETP effluent, implementing the novel technologies would have no significant impact on the water quality for irrigation and would not impact the risk to farmers. This study did also not consider other potential exposure routes for farmers, such as soil contact, which are nonetheless important for a comprehensive assessment of health risks related to Cr.

5. Conclusions

The total Cr levels in the STP's influent were extremely high, and proving that tanneries were informally discharged into the sewers. The concentrations of Cr(VI) in the wastewater, the most hazardous Cr oxidation state, were below 100 µg/L. The total Cr concentrations in the influent ranged from 2.49 mg/L up to 14.95 mg/L. Cr was mainly present in the influent as Cr(III) and Cr(III) concentrations in the conventional and novel technologies' effluents were below the effluent discharge standards of 2 mg/L. Cr accumulated in the primary sludge with concentrations of total Cr ranging from 8,672 to 17,525 mg/kg_{DM} , which consisted mainly of Cr(III), although currently classified as less hazardous it has been associated with adverse health effects in tannery workers. The presence of Cr(VI) levels in the primary sludge raises concerns about its disposal and reuse. The recirculation of the activated sludge led to an accumulation of Cr within the aeration tank, which will pose an occupational risk for workers and affects the secondary treatment's efficiency. The use of the mass balance approach allowed for the exploration of the fate of Cr under normal operating conditions, which would not have been possible through concentration data alone.

Ten health risks related to Cr exposure were identified for the STP workers using the conventional treatment, including three high-risks for handling the primary sludge. The most frequent exposure route was through skin contact. The theoretical impact of implementing the novel technologies would reduce Cr concentrations in the effluent by up to six times. These technologies also reduce direct contact with untreated sewage and decrease sludge production, thereby lowering the frequency of sludge handling risks and, as a consequence, the overall risk score for secondary treatment. However, since most Cr is accumulated in the primary sludge, it would not reduce the major health risks associated with sludge management for STP workers. For Cr, the health risk focus should shift from the overall treatment process to sludge management, as 80% of the total Cr is accumulated in the sludge. Future technical interventions in India should prioritize sludge treatment and disposal strategies rather than focus on effluent.

Farmers using mixed effluent from STP and CETP are at high-risk due to direct skin contact during flood irrigation and general farming activities. The Cr concentration in the mixed effluent exceeds MRLs for skin contact exposure, leading to a high-risk score for these activities. The CETP does not comply with the maximum permissible discharge limit for Cr (2 mg/L) (MoEFCC, 2016). A key finding is that the farmers' risk related to Cr exposure predominantly came from the CETP. Due to this, no significant improvement in the health risks will occur for the farmers reusing the effluent if the novel technologies were implemented. An additional risk assessment is required for sludge reuse amongst farmers and municipal gardeners, but this was beyond the scope of this study.

This study highlights the amount of Cr being discharged into the environment in Kanpur. It is therefore crucial that further research is conducted to explore the fate, health effects, and environmental impact of Cr in this area. Even though Cr(VI) levels were below 100 μg/L, effects on aquatic and terrestrial biota should be further investigated. Additionally, since the study was carried out during the monsoon season, it acknowledges the possibility of seasonal variations in Cr levels, which may pose additional hazards. Therefore, further studies should investigate these seasonal patterns and their potential exposure routes and health risks.

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Institutional review board statement

This study was conducted following the research protocols and guidelines of the Declaration approved by IHE Delft Research Ethics Committee (Approval Number: IHE-RECO-2023-kce001bwa03, Date: 2023-07-19).

Informed consent statement

Informed consent was obtained from all key informants involved in the study.

Author contributions

Conceptualization, C.F; Data curation, K.M.C.V.; Formal analysis, K.M.C.V., C.F, L.B; Funding acquisition, C.M.H.; Investigation, K.M.C.V.; Methodology, K.M.C.V., C.F., L.B., C.M.H.; Project administration, C.M.H.; Supervision, C.F., L.B., C.M.H.; Visualization, K.M.C.V., C.F.; Writing—original draft, K.M.C.V., C.F., L.B.; Writing—review and editing, CF., K.M.C.V., L.B., C.M.H.

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Disclosure statement

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Data availability statement

The data presented in this study is available within the article and Supplementary Material to this article.

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